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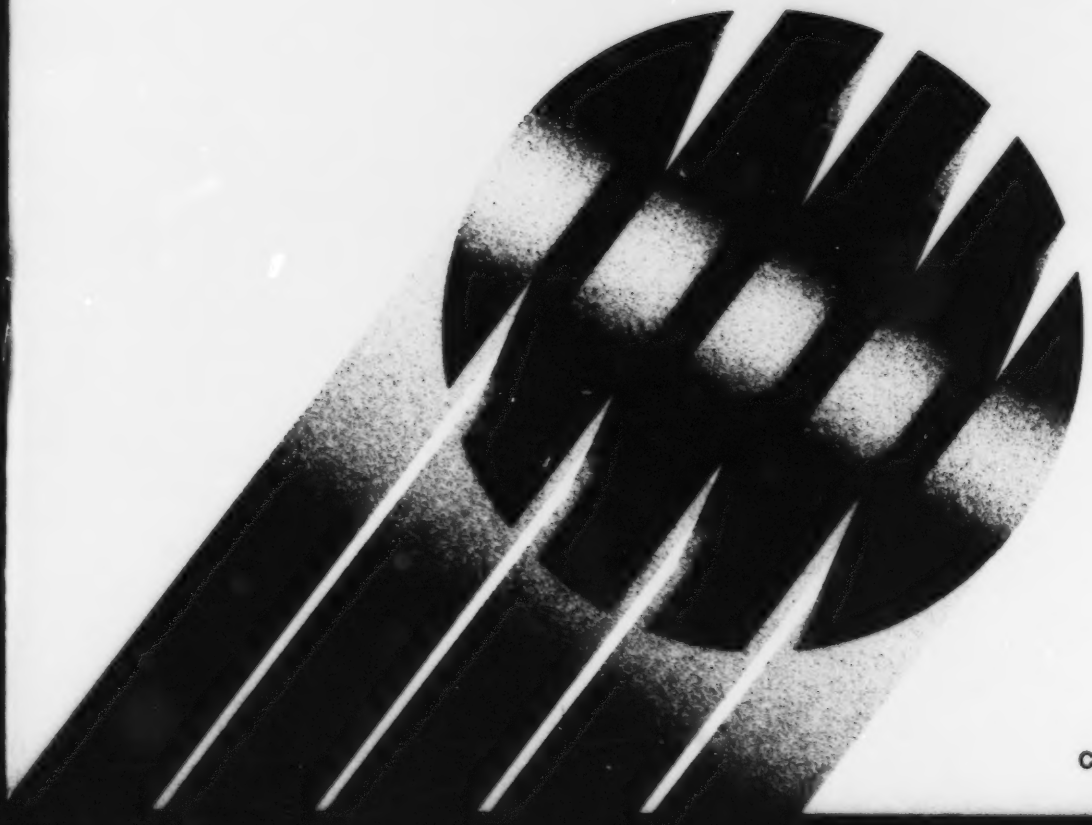
Mineralogical and Image Analysis Study of Artificially Weathered Tailings from the Midwest Uranium Ore in Saskatchewan

W. Petruk and R.G. Pinard
Mineral Sciences Laboratories



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MINERALOGICAL AND IMAGE ANALYSIS STUDY OF ARTIFICIALLY WEATHERED TAILINGS FROM THE MIDWEST URANIUM ORE IN SASKATCHEWAN

by

W. Petruk* and R.G. Pinard**

Abstract

A mineralogical and image analysis study was conducted on samples of U tailings from pilot-plant runs on Midwest ore in Northern Saskatchewan. The tailings had been subjected to simulated weathering for the equivalent of 10 years in a tailings facility (lysimeter); the pH varied from 7.0 at the beginning of the test to 5.9 at the end. The results show that during the simulated weathering some of the gersdorffite, uraninite, galena and pyrite had oxidized, with gersdorffite being the most reactive and pyrite the least. Reaction was evident at the top of the lysimeter, but was most intense at the seepage discharge. Some of the released ions precipitated as Ni, Pb, and U arsenates, sulpharsenates, sulphates and oxides on particles of all minerals. Green crystals of Ni sulphate and white crystals of Ca sulphate had precipitated at the surface of the lysimeter. Clay minerals and chlorite had washed from the top of the lysimeter towards the seepage discharge.

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ÉTUDE MINÉRALOGIQUE ET PAR ANALYSE D'IMAGE DE RÉSIDUS AYANT SUBI UNE MÉTÉORISATION ARTIFICIELLE ET PROVENANT DE MINERAI D'URANIUM DU MIDWEST DE LA SASKATCHEWAN

par

W. Petruk* et R.G. Pinard**

Résumé

Une étude minéralogique et par analyse d'image a été effectuée sur des échantillons de résidus d'U issus du traitement en usine pilote de minerai du Midwest provenant du nord de la Saskatchewan. Les résidus avaient été soumis à une météorisation artificielle équivalant à 10 ans dans un bac pour résidus (lysimètre); leur pH est passé de 7 à 5,9 du début à la fin de l'essai. Les résultats montrent que pendant la météorisation artificielle une partie de la gersdorffite, de l'uraninite, de la galène et de la pyrite s'est oxydée, la gersdorffite ayant été la plus réactive et la pyrite la moins réactive. La réaction était évidente à la partie supérieure du lysimètre, mais était la plus intense à la décharge de l'infiltration. Certains ions libérés se sont précipités sous forme d'arséniates, de sulfo–arséniates, de sulfates et d'oxydes de Ni, de Pb et d'U sur des particules de tous les minéraux. Des cristaux verts de sulfate de Ni et des cristaux blancs de sulfate de Ca s'étaient précipités à la surface du lysimètre. Des minéraux argileux et de la chlorite ont été entraînés de la partie supérieure du lysimètre vers la décharge.

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INTRODUCTION

The weathering of tailings from ore of the Midwest unconformity-type U deposit in Northern Saskatchewan was investigated as part of a CANMET U tailings project. Simulated weathering was carried out by Ontario Research Foundation (ORF) under a contract from CANMET, and the weathered products were analyzed by a variety of techniques (G.M. Ritcey, oral comm. 1984-87). About 11.5 tonnes of U tailings had been produced by pilot-plant runs on the ore by ORF under separate contracts. The acidic tailings were treated with Ca hydroxide and deposited into a tailings facility (lysimeter) by a layered tailings technique (1). The last deposition was in July 1982. The lysimeter was 7.3 m by 9.1 m by 60 cm high, and was designed to collect both seepage and surface runoff (1). The average composition of the tailings was about 0.25 wt % U, 2.5 wt % As, 1.3 wt % Ni and 4000 pCi/g Ra-266. Northern Saskatchewan weather conditions were simulated over the tailings with a night-plus-day cycle of 4.8 hours; hence, the test was considered to be equivalent to 10 years of weathering. The pH varied from 7.0 at the beginning of the test to 5.9 at the end. Cores were taken periodically, the final cores being taken in spring 1984. Mineralogical analyses were performed on the final cores and on freshly leached tailings to determine changes that had been produced, and thereby to provide data for predicting weathering of tailings and migration of elements. The analysis involved identifying and characterizing the minerals and phases, and determining mineral quantities. The minerals were identified with a microprobe equipped with an energy dispersive X-ray analyzer (EDXA), and the mineral quantities were determined with the MP-SEM-IPS image analysis system (2), which consists of a Kontron SEM-IPS image analyzer interfaced with a Jeol 733 microprobe and a Tracor Northern 2000 EDXA.

SAMPLES

Seven cores were taken from the lysimeter by ORF personnel. The lysimeter surface had been subdivided into 100 units using nine rows from intake to discharge end, labelled B to J, and nine rows from right to left, labelled 1 to 9. The cores were taken from rows B, E, H and J at points 1.5, 5 and 8.5. Hence, they were designated as B-1.5, B-5, B-8.5, E-5, H-1.5, H-8.5 and J-5 (Fig. 1). Samples were taken from the top, middle and bottom of each core. As the size range of the particles in the samples was too wide to perform mineralogical analyses, the samples were split by ORF into +74 μm, 74 to 12 μm and -12 μm fractions. Concentrates of the heavy minerals were prepared in the Process Mineralogy Section, CANMET, from the +74 μm fractions by separating the fractions into sink and float sub-fractions with a heavy liquid having a specific gravity of 3.33. Eighty-four samples were thereby produced from the seven cores. Polished sections were prepared from all samples and analyzed.

MINERALOGY

The minerals were identified by qualitative and quantitative EDXA of grains in polished sections. The quantitative EDXA analyses were performed by analyzing for U, Ni, Fe, Co, Pb, Al, Si, Ca, K, As and S, using CANMET microprobe standards. The difference between total for analyzed value and 100 was assumed to be due to oxygen, OH and H_2O. The presence of Mg was determined qualitatively. The samples and standards were analyzed at 20 kV and 10 nA with a Jeol 733 microprobe interfaced with a Tracor Northern 2000 EDXA. ZAF corrections employing nine elements were used for all analyses. To obtain reliable data, the analyses were performed only on grains that appeared homogeneous in the backscattered electron image (BSE), that were larger than 10 μm in diameter, and that did not decompose under the electron beam.

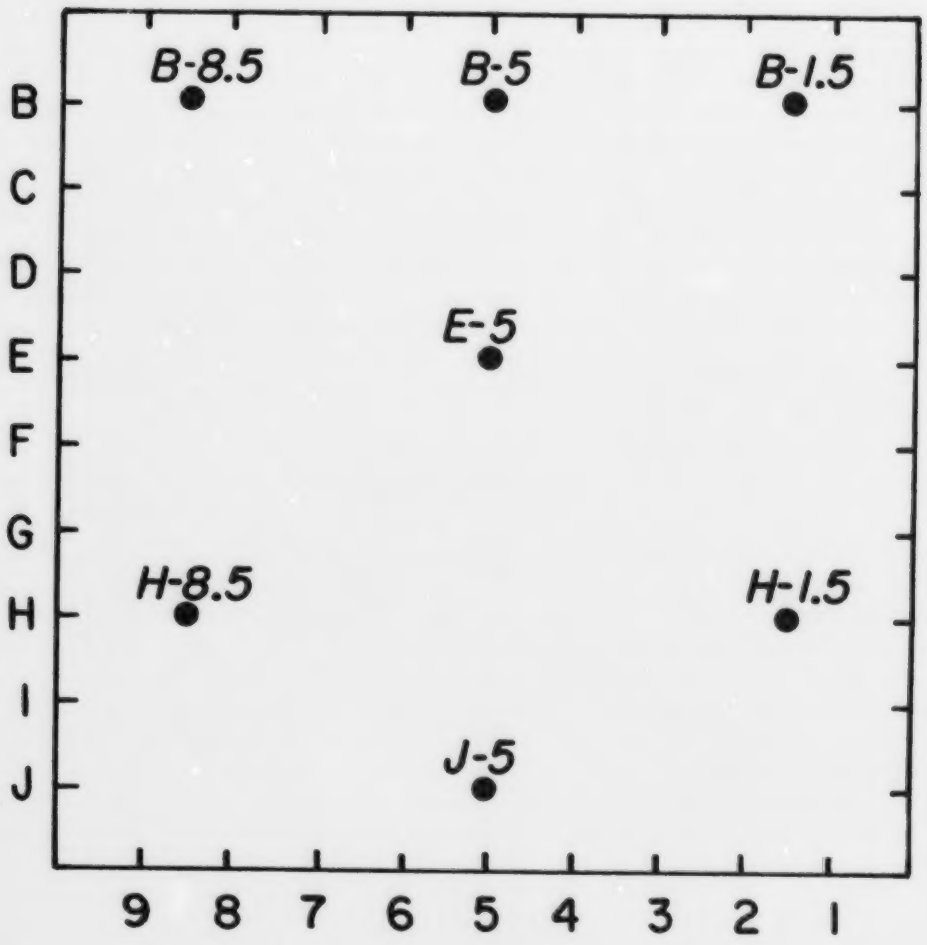

Fig. 1 – Plan of lysimeter showing drill core locations.

Table 1 – Average mineral quantities in sized fractions and combined sample in the lysimeter

Minerals	Total sample	+75 μm 3.33 sink	+74 μm 3.33 float	74 to −12 μm	−12 μm
Silicates	96.1	10.6	–	88.2	–
Quartz + plagioclase	–	–	83.5	–	13.7
K–feldspar + calcite	–	–	6.8	–	14.5
Mafics*	–	–	1.2	–	3.5
Illite	–	–	–	–	41.8
Mica (muscovite)	–	–	7.4	–	13.7
Zircon	0.3	5.4	tr	0.4	0.4
Gypsum	–	–	–	–	0.1
Alteration products (Types 1–3)	1.6	4.9	0.8	1.5	10.0
Alteration product (Type 4)	tr	0.4	–	0.1	–
Rutile	0.3	8.3	0.1	0.8	0.1
Ilmenite	tr	2.7	tr	0.1	–
Hematite + magnetite + goethite	0.2	10.8	0.1	0.8	0.2
Pyrite	0.6	13.3	tr	1.0	0.9
Chalcopyrite	tr	1.5	–	0.3	tr
Sphalerite	tr	4.2	–	0.2	–
Galena	0.3	12.1	0.1	0.6	0.2
Uraninite	0.2	12.8	tr	0.3	tr
Gersdorffite	0.4	13.0	0.1	1.7	0.1
Rammelsbergite	tr	–	–	4.0	–
Nickeline	tr	–	–	–	0.8
Millerite	tr	tr	–	–	–
Total	100.0	100.0	100.0	100.0	100.0

*Mafics = pyroxenes, amphiboles, biotite, chlorite and titanite

+74 μm, 3.33 sink = 0.2 wt % of total sample
+74 μm, 3.33 float = 49.8 wt % of total sample
74 to −12 μm = 16.0 wt % of total sample
−12 μm = 34.0 wt % of total sample

General Mineralogy

The silicate minerals identified in the samples are quartz, K–feldspar, plagioclase, chlorite, biotite, mafic silicate minerals (pyroxenes and/or amphiboles), muscovite, clay (illite), titanite and zircon. The carbonate is calcite. The oxide minerals are rutile, ilmenite, hematite, magnetite, goethite and uraninite. The sulphides, sulpharsenides and arsenides are galena, pyrite, chalcopyrite, sphalerite, millerite, gersdorffite, rammelsbergite and nickeline. The sulphates and arsenates are $CaSO_4.nH_2O$, barite ($BaSO_4$) and Ni, U and Pb arsenates, sulpharsenates and sulphates. An Fe arsenate, determined by X–ray diffraction (XRD), was reported by ORF, but none was found during the current study. It is possible that the Ni arsenate, which is common in the samples, has a similar XRD pattern to Fe arsenate and is identified as Fe arsenate by XRD. On the other hand, Fe arsenate may have been present in the samples but was lost (dissolved or washed away) during sample preparation. Mineral quantities were determined for all samples and the data were combined into average mineral quantities for each fraction and sub–fraction, and into mineral quantities in a hypothetical "total sample" on the basis of wt % of each fraction and sub–fraction (Table 1). The mineral quantities in "total sample" indicate that most of the U (0.25 wt %) is in uraninite. In contrast, only part of the As and Ni is in the primary minerals (gersdorffite, rammelsbergite and nickeline); most is in alteration products.

Silicate Minerals

The silicate minerals are present as separate particles and as lumps of minute grains. Most of the particles are monominerallic, but a few display intergrowths of several silicate minerals and others contain inclusions of oxides, sulphides, sulpharsenides and arsenides. A few particles are coated with clay minerals, arsenates, sulpharsenates, sulphates and the lump material. The lump material consists of very small grains of silicate minerals cemented with $CaSO_4.nH_2O$ and with metal–bearing arsenates, sulpharsenates and sulphates.

The term mafic silicate minerals is used for minerals that contain Si, Fe and/or Mg, and one or more of the following elements: Al, Ca, Na and K, but in different proportions than in biotite, chlorite and muscovite. The last three minerals can be identified by their respective EDXA spectra. Most grains identified as mafic silicate minerals are either pyroxenes or amphiboles. No alteration was observed on either the biotite or mafic silicate minerals; hence, their mode of occurrence was not studied.

The muscovite is present as books of crystals (Fig. 2A) and as very small grains. The mineral is associated with clay minerals, and the muscovite booklets contain clumps of arsenates and sulpharsenates.

The clay minerals consist of Al, Si and K, and contain some Fe, Mg and Ca. They were identified as illite by XRD analysis by ORF. EDXA analyses were performed on a mass of illite that occurs on the edge of a gersdorffite grain and contains minute inclusions of gersdorffite (Fig. 2B), and on an illite inclusion in pyrite (Table 2). Minor amounts of Ni, As and S were detected, probably due to minute inclusions of Ni arsenates, sulpharsenates and sulphates. The illite is present as minute grains, lumps and coatings on other minerals, and as an alteration of K–feldspar. The mineral is commonly mixed with $CaSO_4.nH_2O$ and with metal–bearing arsenates, sulpharsenates and sulphates. It is not known whether the illite was formed in the lysimeter or was originally present in the tailings. However, since it is mixed with arsenates and sulphates that were produced in the lysimeter, the illite must have moved to sites where the reaction products were present or the reaction products moved to sites where the illite was present.

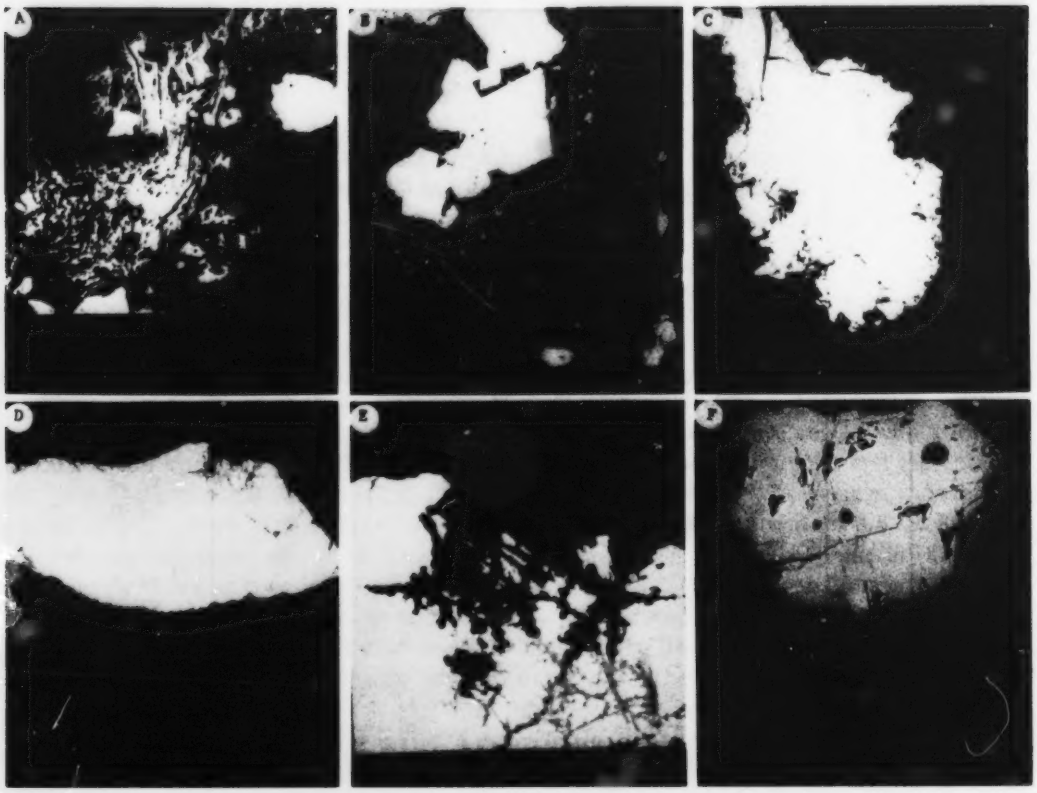

Fig. 2A – Backscattered electron (BSE) photograph of muscovite crystals with inclusions of arsenates and sulpharsenates.

Fig. 2B – BSE photograph showing a large grain of illite (grey) attached to gersdorffite (white) and containing inclusions of gersdorffite. A fibre of tissue covers part of the illite.

Fig. 2C – BSE photograph of uraninite (white) bordered by Type 1(a) alteration product.

Fig. 2D – BSE photograph of uraninite (white) bordered by Type 1(a) alteration product (grey). A Type 2 alteration product coats the Type 1(a) product at the right edge and top of the grain. Type 2 also occurs as a coating on the silicate mineral (grey) at the lower left corner of the photograph.

Fig. 2E – BSE photograph of uraninite (white) with three varieties of veinlets. The light grey veinlets at the centre have the highest U content, 59 wt %; the grey ones towards the edges of the grain contain less U, 46 wt %; and the dark grey parts at the centre of the veinlets contain still less U, 37 wt %.

Fig. 2F – BSE photograph showing galena with a coating of Type 1(a) alteration product.

Table 2 – EDXA analyses of minerals

Mineral	Elements (in wt %)											Remarks
	U	Ni	Fe	Pb	Al	Si	Ca	K	As	S	OH,* 0.4_2	
Illite		1.3	7.0		11.3	20.3		2.9		2.2	54.0	adjacent to gersdorffite
Illite		0.6	5.2		12.7	21.5	0.7	3.8	4.6	0.3	50.6	inclusion in pyrite
Goethite		1.2	57.5		3.0	6.1	0.4		0.8		31.0	edge of pyrite
Uraninite	72.4		0.1	13.8		0.1				0.8	12.8	av of 7 spots**
U-phase 1	59.3			18.5		1.8				1.6	18.8	veinlets at centre of uranium
U-phase 2	45.8	1.9	1.2	31.7		1.3	3.3		6.1	1.6	7.2	veinlets near grain edge
U-phase 3	36.8	2.0	1.5	34.7		1.6	3.2		5.6	2.0	12.6	veinlets at grain edge
Pyrite		2.1	43.5						3.5	51.0		
Millerite		62.0	1.4							36.6		
Altered millerite		31.3	5.2			7.3	1.0			25.2	30.0	
Gersdorffite		33.6							49.5	18.3		
Rammelsbergite		27.5							72.5			
Alteration products												
Type 1(a)	0.5	25.3	0.4			0.7	2.8		28.4	0.1	41.8	av of 11***
Type 1(b)		9.6					2.4		35.4		52.6	on quartz
Type 2	1.7	1.1	2.7	37.5	4.4	1.9	0.6			6.9	43.2	
Type 3(a)	25.8	1.1	1.5	10.2		8.5	2.5			0.8	49.6	on galena
Type 3(b)	8.2	13.2	1.0			2.5	0.8		24.0	12.4	37.8	matrix matrial, av of 6****
Type 4	0.1	28.3	7.6	4.2	0.1	0.3	0.2		1.1	41.1	17.0	av of 4*****

*by difference: **range: U 70.8–75.6, Pb 8.8–16.0, S 0–1.5 wt %; ***range: U 0–1.2, Ni 21.6–30.0, Fe 0–1.8, Si 0.2–1.6, Ca 1.0–4.1, As 23.3–33.4, S 0–1.0 wt %; ****range: U 4.6–12.4, Ni 12.4–14.4, Fe 0.6–1.3, Si 2.0–2.8, Ca 0.6–0.9, As 23.5–24.1, S 11.7–13.2 wt %; *****range: U 0–0.5, Ni 21.6–35.0, Fe 3.9–12.0, Pb 0–10.1, Al 0–0.4, Si 0–0.6, Ca 0.2–0.3, As 0–2.4, S 37.9–42.3 wt %

The chlorite, like illite, is present as minute grains, lumps and coatings on other minerals. The chlorite in the lumps and coatings is mixed with illite, sulphates and arsenates. Hence, like illite, it is at sites where reaction was occurring.

Oxide Minerals

Rutile, ilmenite, hematite and magnetite are present both as separate particles and as inclusions in silicates. No alteration of these minerals was observed; hence, their properties were not studied. The quantities of rutile, ilmenite and hematite plus magnetite were, however, determined (see section on mineral quantities).

The presence of minor amounts of goethite on the edges of some pyrite particles indicates alteration to goethite. Microprobe analysis of goethite that occurs on the edge of a Ni– and As–bearing pyrite particle is given in Table 2.

Uraninite is present in most samples as separate particles, some of which are coated with arsenates, sulpharsenates and sulphates (Fig. 2C and 2D). The boundary between uraninite and the coating is irregular, which suggests that there was reaction at the uraninite surface. The average of seven EDXA analyses of uraninite is, in wt %, U 72.4 and Pb 13.8, range U 70.8–75.6 and Pb 8.8–16.8 (Table 2). The uraninite contains veinlets of three U–bearing phases (Fig. 2E). The phase in veinlets near centres of uraninite grains contains, in wt %, U 59.3 and Pb 18.5 (Table 2); the phase in veinlets towards the edges of uraninite grains contains, in wt %, U 45.8 and Pb 31.7; and the phase in veinlets at the edges of the grains contains, in wt %, U 36.8 and Pb 34.7 (Table 2). The U–bearing veinlets in uraninite are interpreted as original minerals in the ore rather than lysimeter alterations.

Sulphides, Sulpharsenides and Arsenides

The sulphides, sulpharsenides and arsenides are present as separate particles and as inclusions in silicate minerals. Some particles are coated with arsenates, sulpharsenates, sulphates and clay minerals. The boundary between the grain surface and coating is smooth for some grains (Fig. 2F) and corroded for others (Fig. 3A). The corroded grain boundaries indicate reactions at the grain interface.

Only small amounts of sphalerite and chalcopyrite are present, but a few grains are coated with arsenates and sulpharsenates. The presence of a sharp boundary between the mineral and coating is interpreted to indicate that there is no appreciable reaction at the mineral surface, and that the coating, in essence, represents a depositional phase rather than an in–situ reaction product.

Galena is present in most samples as separate particles and as inclusions in silicate minerals. Some galena particles are coated with arsenates, sulpharsenates, sulphates and U–Pb oxides (Fig. 2F and 3A), and the grain boundary between the galena and coating is corroded in some places. The corrosion indicates that there is some reaction at the grain surfaces of galena.

The pyrite also occurs as separate particles and as inclusions in silicate minerals. A few particles are coated with arsenates, sulphates, clay minerals and goethite. The grain boundaries between the pyrite and coatings are sharp (Fig. 3B) to slightly corroded (Fig. 3C), which indicates that very little reaction took place on the pyrite grain surfaces. Compositions obtained from most of the pyrite grains give about 2 wt % Ni and 3 wt % As (Table 2), and higher amounts in some places (Fig. 3D and 3E). The pyrite grain boundaries in some pyrite particles are delineated (Fig. 3F and 4A) to an unusual

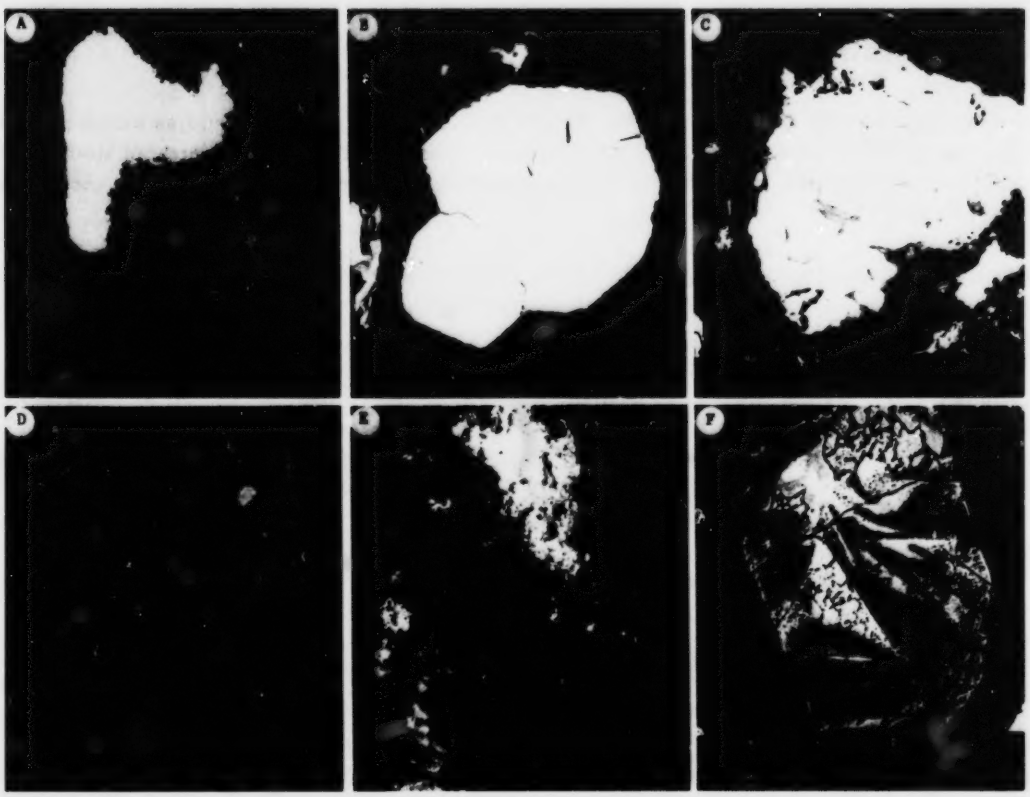

Fig. 3A – BSE photograph of galena with a coating of Type 1(a) alteration product (note irregular boundary between galena and alteration product).

Fig. 3B – BSE photograph of pyrite with a coating of Type 1(a) alteration product (note sharp boundary between pyrite and coating).

Fig. 3C – BSE photograph of pyrite with a coating of both Type 1(a) alteration product and goethite (not differentiated in photograph). The boundary between pyrite and goethite is irregular.

Fig. 3D – BSE photograph of a pyrite grain containing 2 wt % Ni and 3 wt % As. The darkest parts of the grain do not contain Ni or As, and the white parts have higher As contents.

Fig. 3E – BSE photograph of a zoned pyrite grain. The white areas are pyrite that is enriched in As and contains detectable (by EDXA) amounts of U. The pyrite in the grey areas contains detectable As, whereas the pyrite in the dark grey area does not.

Fig. 3F – BSE photograph of pyrite with delineated grain boundaries.

degree. The delineation could have occurred by alteration in the lysimeter, or it may be due to the occurrence of pyrite in a U–bearing orebody. EDXA analyses indicate that the spaces between the pyrite grains contain trace amounts of illite. A minor amount of the pyrite at the particle edges has altered to goethite.

A few clusters of prismatic millerite grains were found (Table 2). The millerite has been either partly replaced by or altered to a Ni sulphate (Fig. 4B and Table 2). It is possible that Si detected in the Ni sulphate is due to impurities, although this is difficult to ascertain since the grains are small and too poorly polished to obtain good analyses.

Gersdorffite is the most common As–bearing mineral in the samples. It occurs as separate particles and as inclusions in silicate minerals. Some particles are bordered by layers of arsenates, sulpharsenates, sulphates and clay minerals. Many of the gersdorffite grain edges are corroded by Ni arsenates, and remnants of gersdorffite are present in the Ni arsenate coatings (Fig. 4C). In addition, minute inclusions of gersdorffite are present in massive illite that is adjacent to large gersdorffite grains (Fig. 2B). EDXA analyses indicate that the gersdorffite is slightly As–rich (Table 2).

A few grains of rammelsbergite and nickeline were found. Some are coated with Ni arsenates but it was not determined whether the rammelsbergite and nickeline have been altered.

Sulphates, Arsenates and Sulpharsenates

Ca sulphate is common in the samples. It is present as minute grains associated with very fine grained particles of silicate minerals, commonly as a cementing medium. A few white crystals were precipitated on the surface of the material in the lysimeter. Semiquantitative spectrographic analyses by ORF (1) show that they are Ca sulphate. A minor amount of barite, identified by XRD, was found as lumps of greenish material at the surface of the lysimeter.

Arsenates, sulpharsenates and sulphates were formed by oxidation of gersdorffite, uraninite, galena and pyrite, and subsequent precipitation of released ions as coatings or layers on most mineral grains, particularly on gersdorffite and uraninite. The coatings (alteration products) were classified into four types on the basis of composition.

Type 1(a) is the most common alteration product. It occurs as a coating on every mineral (Fig. 2C, 2D, 2F, 3A, 3B, 3C, 4C and 4D), but only a few particles of each mineral are coated. The average composition, determined by analyzing 11 grains with quantitative EDXA, is reported in Table 2. The Ni:As ratio (in atomic proportions) is about 1:1, and the phase contains a minor amount of Ca and trace amounts of Si, U, Fe and S. The phase is, therefore, a *Ni arsenate*, probably hydrous. In a few places the outer edge of Type 1(a) coating on gersdorffite is enriched about 5 wt % in As, and depleted about 5 wt % in Ni.

Type 1(b) alteration product is also a Ni arsenic phase (Table 2), but the Ni:As ratio (in atomic proportions) is 1:3; hence, it is a *Ni triarsenate*, probably hydrous. The Ni triarsenate was found only as a coating on quartz.

Type 2 alteration product occurs as an outer layer on Type 1(a) coating on uraninite and gersdorffite (Fig. 4D), and as a coating on silicate grains. EDXA analyses show that it is a Pb sulphate that contains some Fe, Si, Al, U, Ni, Ca and As (Table 2), and XRD analysis gives the pattern of Pb sulphate (anglesite).

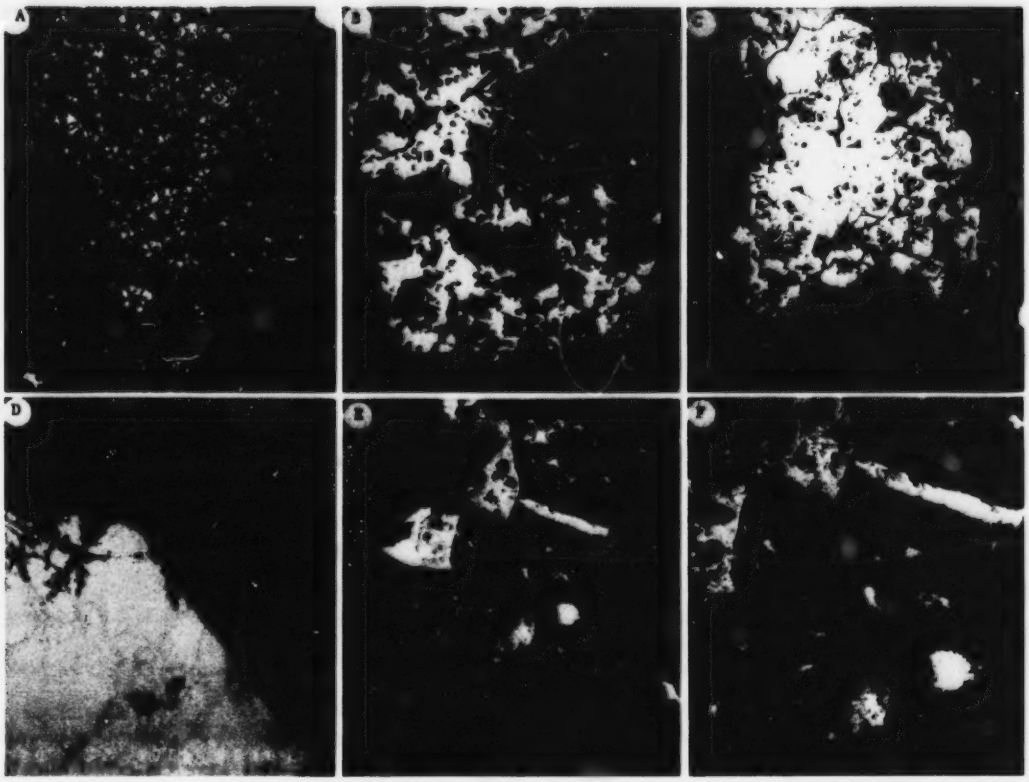

Fig. 4A – BSE photograph of pyrite with delineated grain boundaries.

Fig. 4B – BSE photograph of millerite coated or partly replaced by Ni sulphate.

Fig. 4C – BSE photograph of gersdorffite bordered by Type 1(a) alteration product. Note irregular boundary between gersdorffite and alteration product.

Fig. 4D – BSE photograph of uraninite bordered by a layer of Type 1(a) alteration product, which is in turn bordered by Type 2 alteration product.

Fig. 4E – BSE photograph of several particles cemented by Type 3(b) alteration product into a large particle. The large particle has a border of Type 4 alteration product (Ni sulphate) at the right side.

Fig. 4F – High magnification of part of the particle shown in Figure 4E. The inclusions in Type 3 alteration product are bordered by Type 1(a) alteration product.

Type 3(a) alteration product, found as a coating on gersdorffite, is a U–Pb–Si oxide with minor amounts of Ca, Fe, Ni and S (Table 2).

Type 3(b) alteration product is also U–bearing, but is a U–Ni sulpharsenate. It occurs as matrix material between gersdorffite, galena, pyrite, silicate minerals, etc. (Fig. 4F). Some of the gersdorffite and galena inclusions in the Type 3(b) alteration product are coated with Types 1(a) and 2 alteration products (Fig. 4F).

Type 4 alteration product occurs as a coating on particles composed of Type 3(b) alteration product (Fig. 4E), on lumps of clay minerals, and as separate grains. The average composition from four EDXA analyses shows that the Type 4 alteration product is a Ni–Fe sulphate with impurities of Pb, As, Si, Ca, Al and U (Table 2). The Ni+Fe:S ratio (in atomic proportions) is approximately 1:2. Green crystals occasionally develop on the surface of the material in the lysimeter. Semiquantitative spectrographic analyses by ORF (1) show that the major elements are Ni and S; hence, the green crystals are Type 4 alteration product.

Interpretations of Mineral Reactions in Lysimeter

The mineralogical study suggests that gersdorffite, uraninite, galena and pyrite reacted in the lysimeter, and that some of the released ions precipitated. Textural analyses indicate that a Ni arsenate [Type 1(a) alteration product] precipitated first, and was followed by a Ni triarsenate [Type 1(b)]. A Pb sulphate (Type 2) precipitated next, and was followed by a U–Pb–Si oxide [Type 3(a)] and a U–Ni sulpharsenate [Type 3(b)]. The final precipitate was a Ni sulphate (Type 4).

It is interpreted, from the mineralogical observations, that gersdorffite reacted first and released Ni, As and S. The Ni and As were precipitated on all particles as coatings of Ni arsenates and triarsenates. It is probable that the released S acidified the water to a small degree (from pH 7 to 5.9), which in turn reacted with galena and uraninite. Some of the alteration products remained in situ, but most of the released ions were transported and either precipitated as alteration products on other particles or washed away in the discharge water. The released Pb precipitated on all particles as a coating of Pb sulphate. On the other hand, the released U precipitated as a U–Pb–Si oxide, which coated all particles, and as a U–Ni sulpharsenate, which cemented adjacent particles. The remaining Ni precipitated as Ni–Fe sulphate. With the slightly increasing acidity, the pyrite began to oxidize and release Fe; some of the Fe precipitated as goethite.

MINERAL QUANTITIES

Mineral quantities were determined with the MP–SEM–IPS image analysis system (2) to assess (a) the degree of reactivity of gersdorffite, uraninite, galena and pyrite, (b) the extent of precipitation of arsenates, sulpharsenates, sulphates and U–Pb–Si oxides, and (c) the movement of the minute grains, particularly clay minerals, by the washing effect in the lysimeter. The screened fractions and the sink and float sub–fractions were analyzed (Tables 3, 4, 5 and 6), and the data were combined into data for each sample on the basis of the wt % of each fraction and sub–fraction (Table 7). The results do not show obvious trends for the mineral distributions and suggest that the tailings is a heterogeneous mixture.

A technique that emphasizes anomalies (3) was used to evaluate the mineral quantity data by producing patterns that show zones of relative mineral enrichment and depletion. The technique relates the

Table 3 – Mineral quantities* in +74 micrometre, 3.33 sink sub-fractions

Minerals	B - 1.5			B - 5			B - 8.5			E - 5			H - 1.5			H - 8.5			J - 5			Ave	Sp. Gr
	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot		
Silicates**	3.7	2.0	0.9	1.3	2.4	1.8	8.7	1.9	0.7	0.6	7.6	18.0	2.8	3.2	1.5	1.6	1.9	47.2	4.4	52.7	57.3	10.6	2.9
Zircon	3.1	8.8	4.9	4.1	8.3	2.4	5.0	7.0	10.8	4.9	0.03	2.2	9.0	4.9	7.0	10.2	12.5	3.3	2.8	1.1	0.7	5.4	4.6
Alteration products (Types 1-3)	2.7	5.7	4.3	5.2	7.6	3.1	9.3	8.1	3.9	13.7	7.6	1.8	7.3	3.3	3.9	1.0	1.2	1.2	11.4	0.2	0.8	4.9	2.8
Alteration product (Type 4)	0.5	0.4	0.3	0.1	0.5	0.1	1.5	0.9	0.6	0.5	0.3	0.3	0.2	0.6	0.9	1.0	0.04	0.0	0.0	0.0	0.0	0.4	4.6
Rutile	5.0	9.0	9.1	4.1	9.1	6.6	3.0	16.5	10.6	4.9	10.1	8.8	12.4	8.8	5.2	11.3	26.3	5.1	6.8	0.0	0.4	8.3	4.2
Ilmenite	1.7	2.6	1.1	0.4	1.9	0.1	2.1	2.7	0.7	0.3	0.9	3.2	0.6	1.5	0.6	2.1	1.4	4.6	2.0	13.4	12.6	2.7	4.8
Hematite + magnetite + goethite	11.5	10.3	4.2	5.4	7.9	1.8	15.8	8.9	3.5	3.3	10.7	9.3	7.4	10.2	3.5	7.8	8.0	28.6	8.9	31.4	27.5	10.8	5.5
Pyrite	7.9	29.8	15.1	12.8	18.8	6.8	12.8	22.5	15.1	13.2	22.8	8.1	15.9	10.3	18.3	18.5	22.4	3.9	10.7	0.4	0.3	13.3	5.0
Chalcopyrite	0.5	1.6	2.5	3.1	0.7	1.1	0.7	0.7	1.2	1.1	3.9	1.6	2.8	1.2	1.1	1.3	4.9	0.5	0.3	0.0	0.4	1.5	4.3
Sphalerite	4.8	5.6	3.4	4.7	4.8	2.5	5.3	3.5	5.0	4.4	5.9	4.6	6.4	2.6	5.8	4.4	5.9	1.1	6.9	0.2	0.02	4.2	4.1
Galena	17.9	14.5	5.0	17.4	19.7	10.7	20.9	17.4	14.0	9.2	9.3	2.9	9.7	9.0	19.7	18.1	13.2	0.5	23.0	0.6	0.0	12.1	7.5
Uraninite	36.1	1.8	10.9	35.6	5.3	48.3	13.0	3.5	10.5	18.6	9.3	5.0	11.4	27.6	8.1	4.6	0.7	0.1	16.4	0.0	0.0	12.8	7.5
Gersdorffite	4.6	7.9	38.3	5.8	13.0	14.7	1.9	6.4	23.4	25.3	11.6	34.2	14.1	16.8	24.4	18.1	1.6	3.9	6.4	0.0	0.0	13.0	6.0
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	

*quantities in wt % = areal fraction determined by image analysis, calculated to wt % using specific gravity of minerals
**silicates = quartz, plagioclase, K-feldspar, illite, chlorite, biotite, muscovite, sericite, amphiboles, pyroxene, titanite, and includes calcite and gypsum

Table 4 – Mineral quantities* in +74 micrometre, 3.33 float sub–fractions

Minerals	B - 1.5			B - 5			B - 8.5			E - 5			H - 1.5			H - 8.5			J - 5				Sp.
	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Ave	Gr
Quartz + plagioclase	98.0	96.0	84.6	96.9	95.1	94.2	89.7	94.3	93.4	97.4	94.4	72.0	83.4	90.5	91.7	85.7	90.2	53.9	67.2	51.0	32.2	83.4	2.65
K-feldspar + calcite							4.7	–	–	0.03	0.2	0.5	2.7	4.4	3.0	5.7	4.2	43.1	25.1	47.7	1.1	6.8	2.7
Mafics**	0.3	1.3	1.6	0.9	1.7	3.8		1.1	3.2		–	9.6	–	0.1	0.1	0.1	0.1	0.1	0.2	1.1	0.4	1.2	2.9
Mica (muscovite + sericite)	1.3	2.1	12.2	2.3	2.3	1.4	2.0	3.1	2.1	1.1	4.0	17.4	10.4	4.1	4.1	6.3	4.7	2.5	7.0	0.1	66.0	7.4	2.7
Zircon	–	0.02	0.05	0.17	0.03	–	–	0.09	–	0.2	0.1	0.1	0.1	–	–	–	–	–	–	–	0.01	0.04	4.6
Alteration products + gypsum	0.2	0.4	0.2	0.2	0.5	0.2	2.9	1.3	0.3	0.8	1.1	0.1	3.1	0.6	0.7	1.8	0.6	0.4	0.3	0.02	0.2	0.8	2.8
Rutile	–	0.02	0.04	0.03	0.05	0.01	0.01	0.1	0.3	0.04	0.01	0.03	0.01	0.02	0.1	0.1	0.1	0.01	0.1	0.1	0.1	0.1	4.2
Ilmenite	–	–	–	0.01	–	–	–	–	–	0.04	–	–	–	0.01	–	0.01	–	0.02	0.01	–	–	0.01	4.8
Hematite + magnetite + goethite	–	0.03	–	–	0.01	0.1	0.01	–	0.01	–	–	0.2	0.2	0.2	0.04	–	0.01	0.01	0.01	0.01	0.01	.04	5.5
Pyrite	–	–	0.03	.1	–	0.01	0.01	–	0.02	–	0.01	0.02	0.02	–	0.03	–	0.01	0.01	0.01	0.03	0.01	0.02	5.0
Galena + sphal. + chalcopyrite	0.01	0.06	0.1	0.2	0.1	0.04	0.6	–	–	0.2	0.1	0.02	0.05	0.03	0.1	0.2	0.04	–	0.02	–	0.02	0.1	6.0
Uraninite	0.05	–	–	0.01	–	0.1	–	–	0.2	–	–	0.1		0.03	0.1	–	–	–	–	–	–	0	7.5
Gersdorffite	0.1	0.1	1.2	0.2	0.2	0.2	0.04	–	0.5	0.2	0.05	0.01	0.01	0.02	0.01	0.1	0.04	0.01	0.06	–	–	0.10	6.0
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	

*quantities in wt % = areal fraction determined by image analysis, and calculated to wt % using specific gravities

**mafics = pyroxene, amphiboles, biotite, chlorite and titanite

Table 5 – Mineral quantities* in 12–74 micrometre fractions

Minerals	B – 1.5			B – 5			B – 8.5			E – 5			H – 1.5			H – 8.5			J – 5			Ave	Sp. Gr
	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot		
Silicates**	84.9	84.2	75.2	74.2	75.8	82.4	87.6	93.9	72.7	86.8	90.5	88.7	93.8	93.8	93.6	95.6	95.6	95.4	95.9	95.1	96.5	88.2	7.7
Zircon	0.8	0.5	0.9	0.2	0.5	1.6	1.3	0.3	0.3	0.2	0.5	0.3	0.04	0.1	0.05	0.2	1.0	0.2	0.3	0.1	0.1	0.5	4.6
Alteration products (Types 1-3)	1.7	2.8	1.4	1.8	2.0	0.6	2.6	1.1	2.0	4.5	1.4	0.8	1.8	0.3	0.6	1.6	1.1	0.3	1.0	1.6	1.4	1.5	2.8
Alteration product (Type 4)	0.1	–	0.1	0.02	–	–	–	0.02	–	0.1	0.05	0.01	0.1	0.03	0.03	0.1	0.05	0.2	–	0.01	–	0.04	4.6
Rutile	0.9	0.3	0.5	0.2	0.9	0.04	0.9	1.2	0.8	1.2	1.6	1.5	0.9	0.7	1.6	0.7	0.5	0.9	0.9	0.6	0.2	0.8	4.2
Ilmenite	0.04	–	–	–	–	–	–	–	–	0.2	0.2	0.2	–	–	–	0.1	0.01	0.1	0.04	0.03	0.5	0.1	4.8
Hematite + magnetite + goethite	1.7	1.2	1.4	0.7	0.6	0.5	0.5	1.3	0.7	0.3	0.9	0.2	0.6	1.4	0.8	0.5	1.0	0.8	0.4	0.8	1.3	0.8	5.5
Pyrite	0.7	1.2	2.0	0.9	2.4	1.2	0.9	0.5	1.6	1.4	1.8	1.1	1.3	0.7	0.5	0.5	0.4	0.5	0.3	0.3	0	1.0	5.0
Chalcopyrite	0.4	0.1	0.8	0.4	0.4	0.4	0.2	0.6	0.2	0.2	0.6	0.3	0.1	0.3	0.5	0.03	0.1	0.6	0.1	0.1	–	0.3	4.3
Sphalerite	0.4	0.4	0.05	0.3	0.2	0.4	0.5	0.1	0.5	0.3	0.1	0.2	0.3	0.05	0.03	0.1	0.1	0.1	0.02	0.08	–	0.2	4.1
Galena	2.2	0.9	0.2	0.9	0.9	1.4	1.1	0.1	1.0	0.9	0.8	0.4	0.6	0.3	0.01	0.1	0.1	0.3	0.2	1.1	0	0.6	7.5
Uraninite	1.4	–	0.6	0.8	–	0.7	–	–	0.2	1.3	0.1	0.02	0.2	0.7	0.2	–	–	0.05	0.1	–	–	0.3	7.5
Gersdorffite	1.5	0.1	6.8	0.6	1.1	8.9	0.3	0.1	1.8	1.5	1.4	6.3	0.1	1.5	2.1	0.5	–	0.6	0.3	–	–	1.7	6.0
Rammelsbergite + Nickeline	3.3	8.3	10.1	19.0	15.2	1.9	4.1	0.8	18.2	1.1	0.01	–	0.2	0.1	–	–	0.03	–	0.4	0.2	–	4.0	5.5
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	

*quantities in wt % = areal fraction determined by image analysis, and calculated to wt % using specific gravities
**silicate minerals = quartz, plagioclase, K-feldspar, muscovite, sericite, illite, chlorite, amphiboles, pyroxene, biotite, titanite and includes calcite and gypsum

Table 6 – Mineral quantities* in 12–74 micrometre fractions

Minerals	B - 1.5			B - 5			B - 8.5			E - 5			H - 1.5			H - 8.5			J - 5			Ave	Sp. Gr
	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot		
Quartz + plagioclase	26.0	30.4	0.02	7.0	15.7	23.8	0.2	0.3	0.2	8.8	19.9	19.6	6.5	0.4	2.7	21.8	17.4	27.6	17.0	24.4	18.4	13.7	2.65
K-feldspar + calcite	6.0	10.3	4.1	21.1	17.6	9.7	2.7	2.2	6.1	25.6	29.1	27.2	0.8	13.1	4.3	16.2	23.2	29.2	32.5	20.6	3.0	14.5	2.60
Mafics	2.2	0.6	0.8	6.4	3.5	2.3	0.4	0.3	1.1	10.8	3.8	1.8	–	5.4	0.9	7.9	6.4	5.7	7.8	2.9	1.7	3.5	2.90
Mica (muscovite)	3.9	2.5	47.4	15.1	18.7	4.5	19.9	19.0	23.2	20.0	12.2	5.2	–	10.4	21.0	19.9	14.0	11.3	11.5	7.3	0.04	13.7	2.9
Zircon	–	–	0.02	0.3	–	0.7	–	0.2	0.4	0.3	–	–	0.9	0.1	3.7	–	0.5	1.5	0.6	–	–	0.4	4.6
Alteration products	13.1	17.6	5.4	12.8	11.7	21.2	10.5	12.4	7.9	8.2	7.6	6.2	19.6	7.3	11.9	10.1	8.1	4.9	5.7	7.5	1.3	10.0	2.8
Rutile	–	–	0.4	–	–	–	0.2	–	0.1	–	–	–	–	0.6	0.4	–		–	–	–	–	0.1	4.2
Hematite + magnetite + goethite	1.4	–	–	0.3	–	–	–	0.6	0.8	–	–	–	–	0.2	0.1	0.3	–	–	0.4	–	0.2	0.2	5.5
Pyrite	–	–	0.03	0.5	1.9	–	0.6	–	–	4.0	–	–	–	0.2	–	0.6	2.1	2.2	1.9	3.9	0.2	0.9	5.0
Galena	0.3	–	0.04	–	–	3.2	–	–	0.04	–	–	0.8	–	–	–	–	0.4	–	0.1	–	–	0.2	7.5
Uraninite	–	–	0.05	–	–	–	–	–	0.1	–	–	–	–	–	–	–	–	–	–	–	–	0.01	7.5
Gersdorffite	0.6	–	0.1	–	–	–	0.3	–	–	–	–	–	–	–	0.7	–	–	–	–	–	–	0.1	6.0
Nickeline	–	–	0.05	–	–	–	9.0	1.7	2.4	–	–	–	2.7	–	–	–	–	–	–	–	–	0.8	7.1
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	
Illite ratio (XRD)	0.7	1.6	1.0	1.9	1.0	0.9	1.0	1.0	1.0	0.9	0.8	0.8	0.8	2.0	0.9	0.9	0.9	0.9	0.9	0.9	0.2		
Chlorite ratio (XRD)	1.0	1.0	1.0	1.5	0.6	0.9	1.0	1.0	1.0	1.2	0.9	0.7	1.2	1.0	0.7	1.2	0.7	0.9	1.4	0.9	0.9		

*quantities in wt % = areal fraction determined by image analysis, and calculated to wt % using specific gravities

Table 7 – Mineral quantities* in samples

Minerals	B - 1.5			B - 5			B - 8.5			E - 5			H - 1.5			H - 8.5			J - 5			
	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Top	Mid	Bot	Ave
Silicates**	86.7	85.6	81.5	86.9	86.5	85.7	81.0	81.3	83.5	81.3	82.7	73.6	44.8	52.5	46.7	66.6	68.8	77.9	73.9	70.8	56.2	74.0
Clay and mica	10.6	11.2	14.8	10.4	10.9	9.9	15.4	16.2	13.2	13.8	14.6	22.4	51.2	44.5	42.9	27.5	26.2	17.6	22.8	26.0	42.0	22.1
Zircon	.1	.1	.1	.2	.1	.3	.2	.1	.2	.2	.1	.1	.3	.1	2.2	.01	.5	.9	.4	.02	.1	0.3
Alteration products	.5	1.3	1.0	.7	.8	.7	1.8	1.6	.9	2.0	1.2	1.6	2.3	1.1	6.2	4.0	2.7	1.7	1.0	.6	.4	1.6
Rutile	.1	.2	.2	.1	.2	.1	.2	.3	.4	.2	.3	.3	.7	.5	.6	.7	.1	.2	.2	.1	.1	.3
Hematite + mag. + ilmen. + goethite	.5	.2	.2	.1	.1	.1	.2	.3	.2	.1	.3	.2	.2	.4	.2	.3	.2	.2	.3	.2	.6	.2
Pyrite***	.2	.2	.2	.3	.7	.2	.3	.1	.3	1.4	.3	.2	.1	.3	.1	.6	1.3	1.3	1.1	2.1	.2	.6
Galena	.5	.2	.2	.8	.3	.8	.7	.1	.2	.3	.2	.3	.1	.1	.1	.2	.2	.1	.1	.2	.4	.3
Uraninite	.5	.01	.7	.3	.03	.8	11.1	.01	.3	.3	.04	.02	.05	.2	.1	.04	-	.01	.05	-	-	.2
Gersdorffite****	.3	.04	1.1	.2	.4	1.4	.1	.01	.8	.4	.3	1.1	.03	.3	.9	.1	.01	.1	.1	-	-	.4
Total	100.5	100.0	100.0	100.0	100.0	100.0	100.0	100.4	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

*quantities in wt % = determined by combining values from Tables 3 to 6 on basis of weight of each fraction
**includes quartz, plagioclase, K-feldspar, chlorite, biotite, amphiboles, pyroxene, titanite, calcite and gypsum
***includes sphalerite, chalcopyrite, millerite, Ni sulphate alteration
****includes rammelsbergite and nickeline

quantity of a mineral in a sample to the average quantity of the mineral in all samples, and to the quantity of all other minerals in the sample. A new number is thereby generated for each mineral in each fraction. The values of the number for the samples from drill cores B–1.5, B–5, B–8.5, E–5, H–1.5, H–8.5 and J–5 were plotted on three plans, top, middle and bottom, and contoured to show zones of enrichment (black) and depletion (radiating lines). The following trends were observed:

1. For the +74 μm, 3.33 sink sub–fractions (Fig. 5), the distributions of ilmenite, Fe oxides, rutile, chalcopyrite, pyrite, sphalerite and zircon are similar. The similarity suggests that the distribution patterns produced by these minerals represent the distributions of the unaltered, +74 μm, heavy minerals in the lysimeter. These results show that, in accordance with the mineralogical observation, pyrite alteration is insignificant.

2. For the +74 μm, 3.33 sink sub–fractions, the distribution patterns for the alteration products (Types 1–3) are similar to the distribution patterns for the unaltered heavy minerals. This similarity is expected since the alteration products precipitated on grains of all minerals.

3. For the +74 μm, 3.33 sink sub–fractions, the distibutions of gersdorffite, uraninite and galena are different from those of the unaltered heavy minerals.

   a. The greatest difference is for gersdorffite, which shows relative depletion at the top, middle and bottom of the cores at the discharge end of the lysimeter. High amounts of gersdorffite remain at the bottom at the intake end.

   b. Uraninite also shows relative depletion at the middle and bottom of the cores at the discharge end. A significant amount of the mineral remains at the bottom of the cores at the intake end.

   c. Some relative depletion of galena is evident at the bottom of the cores at the discharge end, and a high amount remains at the bottom at the intake end.

   These distributions suggest that, under the lysimeter conditions, gersdorffite is the most reactive mineral and, along the entire water course, is partly to completely altered. Uraninite is less reactive and was altered only in the middle and bottom of the lysimeter. Galena was the least reactive and was altered only where all the wash water had collected before discharge.

4. The distributions of minerals in the 12 to 74 μm and +74 μm, 3.33 float sub–fractions do not show any trends.

5. For the −12 μm fraction (Fig. 6), the same distribution was obtained for clay minerals (illite) by EDXA as for illite and chlorite by an independent XRD analysis by ORF. This gives confidence in both sets of analyses.

6. For the −12 μm fraction, the clay minerals, chlorite, quartz, orthoclase, plagioclase and mafic minerals are depleted in the top and middle sections of the cores and enriched at the bottom at the discharge end, presumably because they were washed from the top and trapped at the discharge end.

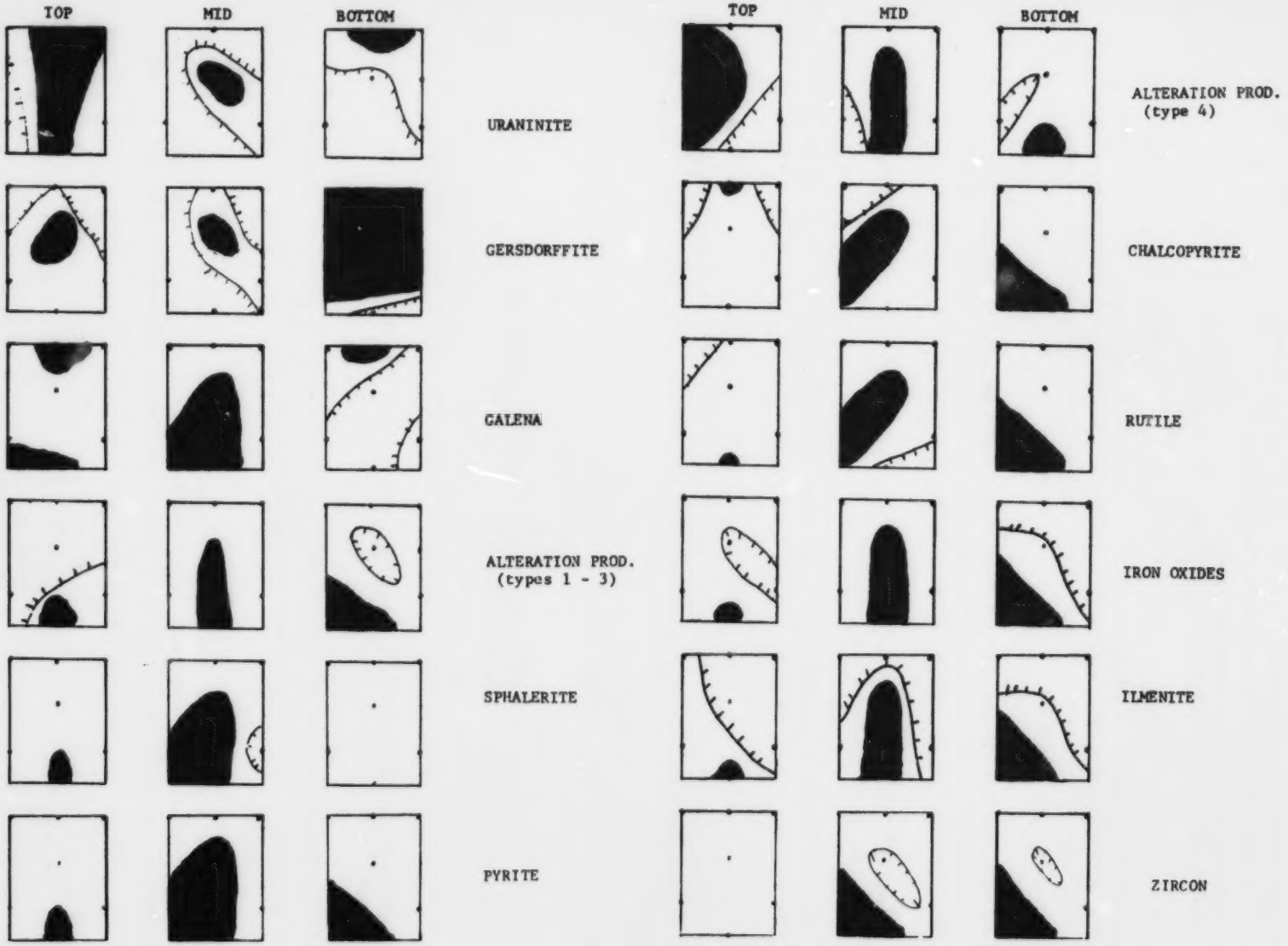

Fig. 5 – Patterns showing enrichment (black) and depletion (radiating lines) for minerals in +74 μm, 3.33 specific gravity sink sub–fractions from samples at the top, middle and bottom of lysimeter.

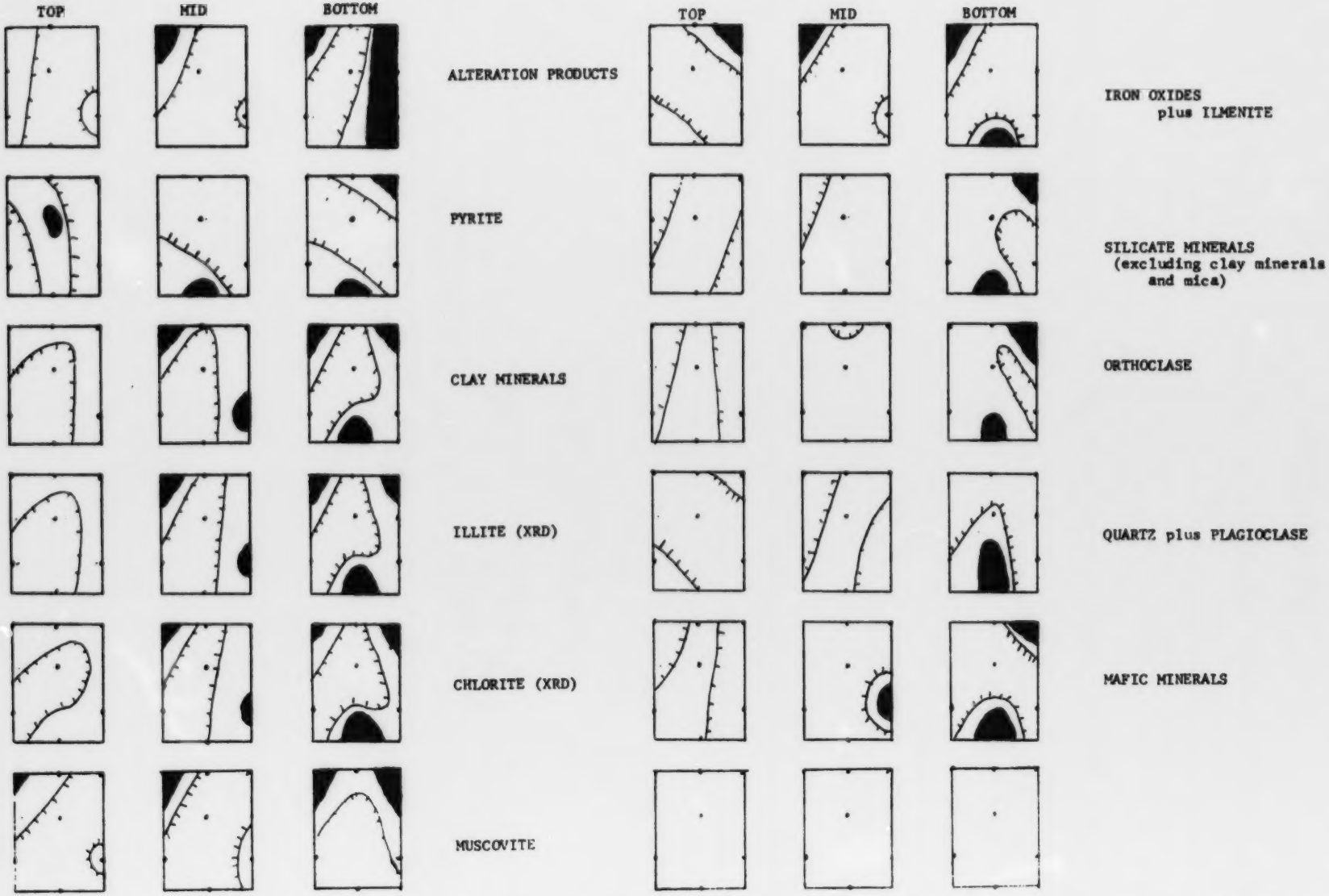

Fig. 6 – Patterns showing enrichment (black) and depletion (radiating lines) for minerals in −12 μm fractions from top, middle and bottom of lysimeter.

CONCLUSIONS

1. The tailings pile in the lysimeter is a heterogeneous mixture of minerals.

2. The distributions of non–reactive heavy minerals are nearly identical. Deviation by a heavy mineral (gersdorffite, uraninite and galena) from the above–noted distribution is interpreted to be caused by the simulated weathering.

3. During simulated weathering, some of the gersdorffite, uraninite and galena, as well as a trace of pyrite, reacted.

4. The released ions precipitated on other minerals as coatings of Ni, U, Pb and Fe arsenates, sulpharsenates, sulphates and oxides.

5. Green crystals of Ni sulphate and white crystals of Ca sulphate precipitated at the surface of the material in the lysimeter.

6. Mineral reactivity, under the conditions in the lysimeter, proceeded in the order gersdorffite, uraninite, galena and pyrite.

7. Mineral reactivity is evident at the top of the lysimeter, but is most intense at the seepage discharge at the bottom because of greatest water circulation.

8. All the gersdorffite and uraninite at the seepage discharge end had reacted, but most of the pyrite had not.

9. Clay minerals and chlorite had washed from the top of the lysimeter towards the seepage discharge.

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